

## INTERNATIONAL PRELIMINARY EXAMINATION REPORT



(PCT Article 36 and Rule 36) Rec'd PCT/PTO 18 MAR 2005

Applicant's or agent's file reference SYN 51075/WO	<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/PEA/416)	
International application No. PCT/GB 03/04109	International filing date (day/month/year) 25.09.2003	Priority date (day/month/year) 25.09.2002
International Patent Classification (IPC) or both national classification and IPC B01J23/75		
Applicant JOHNSON MATTHEY PLC		

- This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
- This REPORT consists of a total of 8 sheets, including this cover sheet.
  - ☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 2 sheets.

- This report contains indications relating to the following items:
  - I ☒ Basis of the opinion
  - II ☐ Priority
  - III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
  - IV ☒ Lack of unity of invention
  - V ☒ Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
  - VI ☐ Certain documents cited
  - VII ☐ Certain defects in the international application
  - VIII ☐ Certain observations on the international application

Date of submission of the demand 22.04.2004	Date of completion of this report 12.01.2005
Name and mailing address of the international preliminary examining authority:  European Patent Office - P.B. 5818 Patentlaan 2 NL-2280 HV Rijswijk - Pays Bas Tel. +31 70 340 - 2040 Tx: 31 651 epo nl Fax: +31 70 340 - 3016	Authorized Officer Zuurdeeg, B Telephone No. +31 70 340-4467 

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/GB 03/04109

**I. Basis of the report**

1. With regard to the **elements** of the international application (*Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17)*):

**Description, Pages**

1-12

as originally filed

**Claims, Numbers**

1-13

filed with telefax on 06.12.2004

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:
- ☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)).

*(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)*

6. Additional observations, if necessary:

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. **PCT/GB 03/04109**

**IV. Lack of unity of invention**

1. In response to the invitation to restrict or pay additional fees, the applicant has:

- ☐ restricted the claims.
- ☐ paid additional fees.
- ☐ paid additional fees under protest.
- ☐ neither restricted nor paid additional fees.

2. ☒ This Authority found that the requirement of unity of invention is not complied with and chose, according to Rule 68.1, not to invite the applicant to restrict or pay additional fees.

3. This Authority considers that the requirement of unity of invention in accordance with Rules 13.1, 13.2 and 13.3 is

- ☐ complied with.
- ☐ not complied with for the following reasons:

4. Consequently, the following parts of the international application were the subject of international preliminary examination in establishing this report:

- ☒ all parts.
- ☐ the parts relating to claims Nos. .

**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

1. Statement

Novelty (N)	Yes: Claims	1-8
	No: Claims	9-13
Inventive step (IS)	Yes: Claims	1-8
	No: Claims	9-13
Industrial applicability (IA)	Yes: Claims	1-13
	No: Claims	

2. Citations and explanations

**see separate sheet**

**Re Item IV**

**Lack of unity of invention**

1. The application lacks unity within the meaning of Rule 13.1 PCT.

The separate inventions or groups of inventions in the application are the following:

- I) Claims 1-9: a process for manufacturing a catalyst, catalyst
  - II) Claim 10: a process for hydrogenation of an organic compound
  - III) Claims 11-12: a process for the formation of a hydrocarbon
  - IV) Claim 13: a process for the oxidation of an organic compound.
2. For any set of independent claims to be unitary, it is essential that they are linked by a common general inventive concept. The present set of claims does not fulfill this requirement in the following respects:
    - 2.1 The application contains independent claims belonging to the following different categories:

- a method for the manufacturing a catalyst (independent claim 1)
- a catalyst (independent claim 9)
- a process for hydrogenation (independent claim 10)
- a process for the formation of a hydrocarbon (independent claim 11)
- a process for oxidation (independent claim 13)

In the present case of claims belonging to different categories, only the catalyst may constitute a link. Therefore, the requirement of unity may only be satisfied under the precondition of the catalyst being **novel and inventive**.

However, the catalyst as defined in claim 9 is already known from R. Oukaci et al., Applied Catalysis A: General 186 (1999) 129-144, see tables 1 and 5; EP-A-1 163 955, see example 3; JP-A-5168932, see examples 3 and 4; and WO 98/47617, see

table 1.

With the catalyst defined in claim 9 being not novel, independent claims 1 and 10 on the one hand and independent claims 10, 11 and 13 on the other hand are not so linked as to form a common general inventive concept.

Hence, the application contains multiple (groups) of inventions (conform item 1) which are not unitary in the meaning of Rule 13.1 PCT.

**Re Item V**

**Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

1. Reference is made to the following documents:  
D1: OUKACI R ET AL: "Comparison of patented Co F-T catalysts using fixed-bed and slurry bubble column reactors" APPLIED CATALYSIS A: GENERAL, ELSEVIER SCIENCE, AMSTERDAM, NL, vol. 186, no. 1-2, 4 October 1999 (1999-10-04), pages 129-144, XP004271930 ISSN: 0926-860X  
D2: EP-A-1 163 955 (KATALEUNA GMBH CATALYSTS) 19 December 2001 (2001-12-19)  
D3: PATENT ABSTRACTS OF JAPAN vol. 017, no. 577 (C-1122), 20 October 1993 (1993-10-20) & JP 05 168932 A (NKK CORP), 2 July 1993 (1993-07-02)  
D4: WO 98/47617 A (EXXON RESEARCH ENGINEERING CO) 29 October 1998 (1998-10-29)  
D5: GB 926 235 A (CHEMETRON CORP) 15 May 1963 (1963-05-15)  
D6: WO 01/62381 A (LOK MARTIN CORNELIS ;ICI PLC (GB); BAILEY STEPHEN (GB); GRAY GAVIN) 30 August 2001 (2001-08-30)
2. The present application does not meet the requirements of Article 33(1) PCT, because the subject-matter of claims 9-13 either is not new in the sense of Article 33(2) PCT or does not involve an inventive step in the sense of Article 33(3) PCT in view of at least one of the documents D1 to D6.

2.1 It is pointed out that a product is not rendered novel merely by the fact that it is

produced by means of a new process. A claim defining a product in terms of a process (i.e. claim 9) is to be construed as a claim to the product as such.

Catalysts comprising cobalt species on a titania support and their use in Fischer-Tropsch synthesis, hydrogenation and oxidation reactions are known, see D1, tables 1, 5; D2, example 3; D3, abstract; D4, table 1.

In his letter of reply, the applicant provided arguments why the subject-matter of claim 9 would be novel over the prior art cited. However, no evidence was confined to this letter to support the statements for novelty, especially in relation to D4. The applicant did not discuss the inventive step of the catalyst claim 9 in view of the prior art.

In principal, the **novelty and inventive step** of the claimed product (according to claim 9) has to be shown over catalysts, which are being prepared by different processes, but have the comparable physical and chemical parameters. The inventive step has to be shown over the whole breadth of scope, i.e. with respect to the temperature range (60 to 110°C) and weight range of cobalt (basically 0<cobalt content<100 wt%), by providing evidence that the surprising effect is obtained over the whole range claimed.

The subject-matter of claims 9-13 is therefore not novel and it does not fulfill the requirements of Article 33(2) PCT, or the subject-matter of claims 9-13 does not involve an inventive step and does not fulfill the requirements of Article 33(3) PCT.

- 2.2 The subject-matter of claims 1-8 is novel over the available prior art documents D1-D6.

Document D1 is considered to represent the closest prior art. The subject-matter of claim 1 differs from D1 in that different processes for preparing cobalt on titania catalysts are disclosed.

The applicant points out that when comparing the catalyst T-0 of table 5 in D1, which contains 12 wt% Co on TiO<sub>2</sub>, with catalysts 8A to 8E, which comprise approximately 13 wt% cobalt and which were prepared according to the process of the current invention, it is clear that the cobalt metal surface of the catalyst disclosed in D1,

which is 12.63 m<sup>2</sup>/g cobalt, based on H<sub>2</sub>-chemisorption data provided in table 6 (19 μmol H<sub>2</sub>/g) and using the formula on pages 5 and 6 of the current specification, is lower than the metal surface areas of the catalysts prepared by the process according to the invention, and which are mentioned in table 2 of the current specification.

However, the application does not contain evidence that this unexpected effect is obtained over the whole range claimed. Alleged but unsupported advantages cannot be taken into consideration in respect of the determination of the problem underlying the application.

The technical problem underlying the invention can be seen as to provide a further process for preparing cobalt on titania catalysts.

The process of heat decomposition precipitation of cobalt ammine carbonate (as defined in claim 1) is already known for silica and alumina supports (see D6 and WO96/04072 (page 1 of the specification)).

However, there is no suggestion in D6 that this method could be employed to make cobalt catalysts supported on titania. Besides, it appears not to be practical to make catalysts containing more than 12 wt% cobalt using P25 support (see table 1 of D1) with the preparation methods used in D1, whereas the examples of the present application demonstrate titania supported catalysts having >26 wt% cobalt on P25 (examples 10A-10C) could be prepared using the method of claim 1, while maintaining a high cobalt metal surface area. The solution provided for in independent claim 1 is not obvious in the light of the prior art.

Thus, an inventive step could be recognised for the subject-matter of independent claim 1 (Article 33(3) PCT).

2.3 Claims 2-8 are dependent on claim 1 and as such also meet the requirements of the PCT with respect to novelty and inventive step.

### 3. Clarity

3.1 It is clear from the description on page 4, third full paragraph ("...amounts **should be** such that...") that the following feature is essential to the definition of the invention:

(1) the pH of the cobalt ammine carbonate solution is in the range 7.5 to 12

Since independent claim 1 does not contain this feature, it does not meet the requirement following from Article 6 PCT taken in combination with Rule 6.3(b) PCT that any independent claim must contain all the technical features essential to the definition of the invention.

4. Certain published documents (Rule 70.10 PCT)

Application No Patent No	Publication date (day/month/year)	Filing date (day/month/year)	Priority date (valid claim) (day/month/year)
WO02/089978	14.11.2002	06.05.2002	08.05.2001
WO03/024905	27.03.2003	23.09.2002	21.09.2001



## **Claims.**

- 1. A process for manufacturing a catalyst which comprises a cobalt species on a titania support, comprising mixing together said titania support and an aqueous solution of cobalt ammine carbonate, and heating to a temperature in the range 60 to 110°C to effect decomposition of the cobalt ammine carbonate and precipitation of an insoluble cobalt compound onto said titania support.**
- 2. A process as claimed in claim 1, comprising saturating a titania support with an aqueous solution of cobalt ammine carbonate, and removing the excess of the solution, before heating the resulting product to a temperature sufficient to effect decomposition of the cobalt ammine carbonate.**
- 3. A process as claimed in claim 1, wherein the mixture of titania support and said cobalt solution is heated to a temperature sufficient to effect decomposition of the cobalt ammine carbonate in situ before separating the solid catalyst from the mixture and drying.**
- 4. A process as claimed in any of claims 1 to 3, wherein the titania support and cobalt solution are maintained at an elevated temperature for a period of at least 60 minutes.**
- 5. A process as claimed in any of claims 1 to 4, further comprising the step of calcining the resulting catalyst product at a temperature between 200 and 600°C.**
- 6. A process as claimed in any of claims 1 to 5 further comprising the step of reducing the resulting catalyst product with hydrogen at a temperature between 300 to 550°C.**
- 7. A process as claimed in claim 6, further comprising the step of dispersing the reduced catalyst in particulate form product in a carrier matrix.**
- 8. A process as claimed in any one of claims 1 to 7, wherein the pH of the mixture of titania particles and aqueous cobalt ammine carbonate complex is maintained above 7.5 during the heating step.**
- 9. A catalyst or catalyst precursor made by the process claimed in any of claims 1 to 8.**

10. A process for the hydrogenation of an organic compound comprising an olefinic, carbonyl, nitrile, nitro or aromatic group, comprising reacting said compound with hydrogen in the presence of a catalyst as claimed in claim 9.
11. A process for the formation of a hydrocarbon by the reaction of carbon monoxide with hydrogen in the presence of a catalyst as claimed in claim 9.
12. A process as claimed in claim 11 or claim 12 further comprising the step of forming an active catalyst in situ by reducing a catalyst precursor as claimed in claim 9 with hydrogen before conducting said hydrogenation reaction.
13. A process for the oxidation of an organic compound by reaction with an oxygen-containing compound in the presence of a catalyst as claimed in claim 9.

EPO - DG 1  
15. 12. 2004

(59)